Chapter 1
Aerosols in the Atmosphere: Sources, Transport, and Multi-decadal Trends

Mian Chin, Thomas Diehl, Huisheng Bian and Tom Kucsera

Abstract We present our recent studies with global modeling and analysis of atmospheric aerosols. We have used the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model and satellite and in situ data to investigate (1) long-term variations of aerosols over polluted and dust source regions and downwind ocean areas in the past three decades and the cause of the changes and (2) anthropogenic and volcanic contributions to the sulfate aerosol in the upper troposphere/lower stratosphere.

1.1 Introduction

Aerosols are liquid or solid particles suspended in the air with typical diameters ranging from a few nanometers (10^{-9} m) to a few tens of micrometers (10^{-6} m). Source of aerosols includes desert and soil dust, wildfire smoke, sea salt particles produced mainly by breaking bubbles in the ocean spray, volcanic eruption, and anthropogenic activities. Major aerosol species in the troposphere are dust, sea salt, black carbon (BC), organic carbon (OC) or sometime called particulate organic matter (POM), sulfate, and nitrate. A large fraction of aerosols is natural in origin, including desert dust, wildfire smoke, sea salt, and volcanic aerosols, whereas

M. Chin (✉) · T. Diehl · H. Bian · T. Kucsera
Atmospheric Chemistry and Dynamics Laboratory, NASA Goddard Space Flight Center, Greenbelt, MD, USA
e-mail: mian.chin@nasa.gov

T. Diehl
Joint Research Center, Ispra, Italy

H. Bian
University of Maryland Baltimore County, Baltimore, MD, USA

T. Kucsera
Universities Space Research Association, Columbia, MD, USA

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others are from human activities, such as fossil fuel combustions from industries and energy use, agriculture land cleaning, and transportation.

Aerosols are removed from the atmosphere by wet and dry depositions that establish average tropospheric aerosol lifetimes at about a few days. Despite their relatively short lifetimes, aerosols originating from one region can travel long distances to affect the environment of downwind regions. This transport demonstrates the hemispheric to global scope of aerosol influences.

Aerosols exert a variety of impacts on the environment. Aerosol particles (aka particulate matter or PM) have long been recognized to affect human health and visibility. Aerosols also have climate effects by interactions with solar and terrestrial radiation and with cloud and precipitation. The net cooling effects caused by aerosols is believed to mask the warming effects caused by greenhouse gases. However, the challenging of quantifying the aerosol climate effects arises from the immerse diversity of aerosols (not only in particle sizes, composition, and origin, but also in the highly variable spatial and temporal distributions), the multifaceted atmospheric processes of aerosol formation and removal, and the complex physical, chemical, and optical properties of aerosol particles. One consequence of this heterogeneity is that the aerosol impacts must be understood and quantified on a regional rather than just a global-average basis.

Anthropogenic activities have caused considerable changes in aerosol composition and loading. Historical emission inventories have estimated that trends in anthropogenic emission are closely tied to economic growth, population density, and technological development, which may explain the regional aerosol variability shown in the long-term observation records. For example, aerosol optical depth (AOD) trends from the satellite and surface-based observations suggest that in the past three decades, the amount of anthropogenic aerosol has de-creased over North America and Europe due to enforced emission control policy, but has increased over East and South Asia due to the fast economic development. This increase of Asian pollution in the past decade was suggested to be responsible to cause the observed increasing trends of aerosol extinction in the stratosphere via the efficient Asian monsoon convective transport; however, the quantification is difficult because of the frequent injection of volcanic clouds to the upper troposphere and lower stratosphere (UTLS) that could overwhelm the signal of Asian pollution in the stratosphere.

Global chemistry transport models are essential tools to integrate our current knowledge of emissions and atmospheric processes of aerosols, analyze remote sensing and in situ observations, and assess the impact in the past, present, and future climate scenarios. We present here our recent studies using the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model and observations on (1) multi-decadal variations of aerosol concentrations and AOD over different regions and (2) anthropogenic and volcanic contributions to the stratospheric aerosols. We will first introduce the GOCART model simulations of atmospheric aerosols and describe the observation datasets, then present the results and analysis, followed by conclusions.

1.2 GOCART Model Simulation of Aerosols in the Atmosphere

The GOCART model is a chemistry transport model driven by the assimilated meteorological fields from the NASA Modern Era Reanalysis for Research and Applications (MERRA). The horizontal resolution of model simulation in this work is at 1.25° longitude by 1° latitude (or 2.5° by 2° for the 30-year simulation) and 72 vertical levels from the surface to 0.01 hPa. Aerosol simulations in GOCART include sulfate, dust, BC, OC, and sea-salt, and the sulfate precursor gas species of SO_2 and dimethyl sulfide (DMS). The "building blocks" of the model include emissions from anthropogenic and natural sources, advection, shallow and deep convections, dry deposition, wet scavenging, and chemical transformation. Aerosol particle sizes are parameterized as a function of relative humidity. Aerosol optical properties, such as extinction, absorption, backscattering, are calculated from the refractive indices and size distributions, both depending on the type of aerosols and wavelengths.

Emissions of SO_2, BC, and OC from fossil fuel and biofuel combustions and biomass burning are taken from the existing emission inventories (A2-ACCMIP, Diehl et al. 2012 or more recent HTAP emission, http://htap.org). Volcanic SO_2 emissions are taken from a recent compilation that includes emission amounts and plume heights based on the volcanic activity database from the Smithsonian Institution's Global Volcanism Program, the satellite observations of SO_2 from the Total Ozone Mapping Spectrometer (TOMS) and the Ozone Monitoring Instrument (OMI), and ancillary information from other observations reported in the literature (Diehl et al. 2012). Dust emissions are calculated as a function of surface topography, surface bareness, 10-m wind speed, and ground wetness (Ginoux et al. 2001). Sea salt and DMS emissions from the ocean are calculated as a function of 10-m wind speed (also the sea water concentration of DMS for calculating DMS emissions). Biogenic source of OC is parameterized as a fraction of monoterpene emission. More detailed description of GOCART model can be found in our previous publications (Chin et al. 2002, 2009, 2014 and references therein.).

1.3 Observations

Aerosol data products from remote sensing and in situ measurements, together with associated time periods and measured quantities used in this study are listed in Table 1.1. They include AOD from satellite observations by AVHRR, TOMS, SeaWiFS, MISR, MODIS, UTLS aerosol extinction profile from SCHIAMACHY and CALIOP, column SO_2 from OMI, and surface concentration from the IMPROVE, EMEP, and University of Miami networks and in the Arctic. All data used in this study are monthly averages.

Table 1.1 Observation data used in this study

Dataset	Location	Period
Satellite AOD		
AVHRR-CDR	Global, ocean	1981–2009
AVHRR-GACP	Global, ocean	1981–2005
TOMS	Global, land + ocean	1980–1992, 1996–2001
SeaWiFS	Global, land + ocean	1997–2009
MISR	Global, land + ocean	2000–2009
MODIS-Terra	Global, land + ocean	2000–2009
MODIS-Aqua	Global, land + ocean	2002–2009
Satellite SO_2 and UTLS aerosol extinction profile		
OMI SO_2	Global	2004–2009
SCHIAMACHY ext.	UTLS	2002–2009
CALIOP ext.	UTLS	2006–2009
Surface aerosol species concentrations		
IMPROVE	U.S.	1988–2007
EMEP	Europe	1980–2008
Arctic	Arctic	1980–2009
Univ. Miami	Islands or coast	1980–2009

1.4 Multi-decadal Aerosol Variations Over Land and Ocean Regions

We divide the globe into 15 land, 12 ocean, and 2 polar regions in our analysis, although in this talk we focus mostly on the NH major pollution source regions, namely USA, Europe, East Asia, and South Asia, the major dust regions of Sahara and Sahel, and their immediate downwind oceanic regions. Note that East Asia and South Asia also contain considerable dust source areas. To investigate the regional trends, we first compare the AOD time series from satellite retrievals and model simulations over these regions (unfortunately the reliable satellite retrievals over land are only available after 1998). The most pronounced feature during the 30-year time period is the worldwide influence of large volcanic eruptions of El Chichón in 1982 and Pinatubo in 1991; sulfate aerosols formed from these big volcanic eruptions can last for several years. For the periods lacking major volcanic influence (i.e., in the mid- to late 1980s and in the 2000s), there is a decreasing AOD trend over the pollution source regions of USA and Europe, and the model shows that such a decrease is mostly driven by the decline of aerosol from combustion AOD, in line with the fossil fuel/biofuel emission reduction trends in these regions. The reduction over Europe is particularly remarkable with the total and combustion AOD in the late 2000s are only about half and one-third of their corresponding values in the early 1980s from. In contrast, over East and South Asia the

combustion AOD has increased significantly by 40 and 120 %, respectively, for the same period of time. However, the overall increase of total AOD is less than 20 % in East and South Asia because of the relatively large amount of dust aerosols in these regions that either have negligible trend (in East Asia) or a decreasing trend (in South Asia) to mask the increasing trend of anthropogenic AOD. The decrease of aerosols over the US and Europe is further corroborated by the long-term surface concentrations measurements from the IMPROVE and EMEP networks.

Over the dust source regions, the model shows a general decreasing trend of dust AOD in Sahara and Sahel since the mid-1980s, but an upward trend in Middle East in the 2000s. There results are consistent with the satellite observations that are mostly available since 2000. Although there are noticeable differences in satellite data over the ocean, one consistent and persistent feature is the decrease of AOD over the tropical North Atlantic in the past three decades. This weakening of dust transport from North Africa to the North Atlantic is also evident in the long-term surface dust concentration record at Barbados where the dust is predominantly from North Africa via trans-Atlantic transport. The decrease of dust over North Africa and tropical North Atlantic can be attributed to the weakening of surface winds over North Africa that has suppressed the dust emission, and the increase of precipitation over the ocean that has made scavenging of dust more efficient. Such meteorological condition shifts are closely connected to the warming of the sea surface in the past three decades.

Despite significant changes of aerosol levels over many regions, model-calculated global average AOD show little trends over the 30 years, because a global averaged number conceals not only the opposing regional changes but also the large spatial inhomogeneity of aerosols. Therefore, assessing aerosol impacts should be done on regional-scale analysis.

1.5 Anthropogenic and Volcanic Contributions to the Decadal Stratospheric Aerosol Change

The fast economic growth in South and East Asia in recent decades has led to an accelerated increase of pollutant emissions. During the summer monsoon season, deep convection can transport pollutants from the boundary layer to the UTLS, evidently shown in the satellite observations of enhanced CO and aerosol in the UTLS region, which led to an hypothesis that the Asian pollution may have caused the increase of stratospheric aerosol levels (Hofmann et al. 2009). On the other hand, volcanic eruptions can inject SO_2 directly into the upper troposphere and stratosphere to produce sulfate aerosols at high altitudes where residence time is much longer, making a disproportionately larger contribution than anthropogenic sources to the aerosol loading in the UTLS compared to their respective emission strengths. To assess the relative contributions of anthropogenic versus volcanic sources to the UTLS aerosols in the last decade, we have performed three model

experiments: (a) base simulation with all emissions, (b) simulation without anthropogenic emissions, and (c) simulation without volcanic emissions. The difference between (a) and (b) is the anthropogenic contribution and between (a) and (c) is the volcanic contribution to the total aerosol loading.

Our base simulations of SO_2 from anthropogenic and volcanic sources are compared with satellite observations from the OMI instrument on the Aura satellite, and UTLS aerosol extinction profiles with the SCHIAMACHY instrument on the European ENVISAT satellite and CALIOP on CALIPSO satellite. The model captures the volcanic events that had large impact on stratospheric aerosol detected by satellite, although discrepancy exists on the absolute magnitude. With the tagged simulations isolating the volcanic and anthropogenic contributions, it is clear that the stratospheric aerosol is overwhelmingly volcanic origin in the past decade (2000–2009). Although the ratio of volcanic/anthropogenic SO_2 emission is only 1:4 on decadal average, the ratio of corresponding stratospheric sulfate amounts is disproportionally 2:1, due to the high altitude injections of volcanic emissions. On the other hand, Asian anthropogenic aerosol does seem to show an increasing trend, in the UTLS region, but its influence is mostly confined in the upper troposphere with well-organized seasonal variations. The model suggests that the volcanic sources could be more responsible than the anthropogenic sources for the apparent increasing trend of stratospheric aerosol in the past decade at certain locations.

1.6 Conclusions

We have presented our recent studies on the decadal to multi-decadal aerosol variations in different regions of the world, estimated the anthropogenic and natural source contributions to these changes, and assess the impact of long-range transport of aerosol from the polluted source regions to the receptor regions. We have found:

Over land regions dominated by pollution aerosols, the 30-year AOD and surface concentration trends are generally consistent with the direction of the regional pollutant emission changes, with aerosol decreasing in USA and Europe but increasing in South Asia and East Asia. Over the Arctic, the model and long-term surface measurements show a significant decrease of sulfate concentrations since the early 1990s, aligned with the decreasing trends over Europe.

There is a general decreasing trend of dust over North Africa and downwind regions in the tropical North Atlantic in the past 30 years, which is driven by the reduction of dust emission in North Africa and an increase of precipitation over the North Atlantic, both likely the results of a North Atlantic Ocean warming trend that has lead to the changes in atmospheric circulation and precipitation.

Despite the significant changes over many regions, model-calculated global averaged AOD values show little trends over land and ocean on a global, annual scale, because increases and decreases in different land regions tend to cancel each other in the global average and little changes occur over large open ocean areas. A globally averaged number conceals not only opposing regional changes but also

the large spatial inhomogeneity of aerosols. This highlights the need for regional-scale analyses, as global average values are not sufficient for assessing aerosol trends and impacts.

Both satellite data and model have shown that even without major explosive volcanic eruptions, volcanic emissions frequently perturb the stratospheric "background" aerosols, making it difficult to define non-volcanic background aerosol values in the stratosphere. Overall, volcanic aerosol is twice as much as the anthropogenic aerosol in the stratosphere in the past decade, which could be more responsible for the decadal increase trend of stratospheric aerosols observed in certain locations.

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Question and Answer

Questioner: Ivanka Stajner

Question: In the comparison for early years AVHRR and TOMS data are used giving different trends over oceans. Are these data of trend-quality? What are the particular retrieval data sets? Can different sensitivity of AVHRR and TOMS to aerosol composition explain different trends?

Answer: These earlier datasets were not designed for detecting aerosols, therefore the retrieval ability and data quality of the aerosol products suffer by several limitations of the instruments. For example, AVHRR does not provide the aerosol retrieval over land and TOMS has to exclude large amount of data that are affected by the clouds. Nonetheless, the AVHRR Climate Data Record dataset we used in this study has been created for the purpose of assessing aerosol trends, albeit only over oceans. TOMS data, on the other hand, are used mainly for features, not for quantitative assessment. The positive oceanic aerosol trends from TOMS is within the uncertainty range of the TOMS data, particularly over the low AOD areas, so it should not be referred as "trends". Over high AOD regions such as high dust and pollution areas, trends from TOMS are consistent with the expected AOD changes.

Questioner: S.T. Rao

Question: From your analysis, what can you say about the aerosol impact on climate from volcanoes versus anthropogenic forcings given different residence times of aerosols from these two sources? How do you attribute aerosols from anthropogenic or volcanic sources?

Answer: Volcanic effects are mostly in the free troposphere and lower stratosphere because of the emission injection heights, in contrast with the near-surface anthropogenic emissions. However, volcanic aerosol have longer lifetime than

anthropogenic aerosol, mainly because they are located far above the surface thus not subject to efficient dry and wet removals. Globally, volcanic forcing is much smaller than the anthropogenic forcing, but in the UTLS it can be the dominant aerosol component. We use the model to separate volcanic and anthropogenic aerosols by "tagging" the emission sources.

Questioner: Jose M. Maldasano

Question: In the results presented with respect to the Middle East area, it has indicated that the AOD had increase in the past ten to fifteen years due to mineral dust, when in that area in recent years has been the Iraq and Syria wars, and also a strong urban and industrial development in all gulf countries, has an explanation for this? Was threshold value has been used to discriminate the AOD due to mineral dust?

Answer: The Middle East AOD increase in the past decade has been observed by satellite data. The Angstrom exponent from the ground-based AERONET observations over the Middle East shows that such an increase is mainly due to the coarse-mode dust aerosols, indicated by the decreasing of the Angstrom exponent in the past decade. Our model simulation supports such findings with our tagged source simulations that show the AOD increase over the Middle East is mainly due to the increase of dust rather than anthropogenic aerosols.

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